

# SCIENCE FOR CERAMICS PRODUCTION

UDC 666.3:666.32

## OPTIMAL MIX FOR BUILDING CERAMICS

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Translated from *Steklo i Keramika*, No. 3, pp. 9–12, March, 2009.

It is determined on the basis of experimental data and calculations that frost-resistant ceramics can be obtained from a mixture of low-melting illite-bearing clays without other, more expensive, refractory clays. Techniques for planning “property – property” experiments were used to formulate the molding mixes. For the optimal content of the molding mixes, a sintered ceramic material with frost-resistance of more than 250 cycles with omni-directional freezing and from 75 to 125 cycles with uni-directional freezing was obtained.

When formulating molding mixes for unglazed tile and facade ceramic articles the precise values of important parameters such as the optimal technological properties of the molding mixes (plasticity, drying and roasting parameters), and frost-resistance must be determined and the articles must have good physical and chemical qualities.

The technological drawbacks of some clay, for example, inadequate plasticity, can be compensated by clay with greater plasticity [1]. Clay containing more of larger clay particles is used to increase the molding-mix plasticity. Often, two or more clays are used for the molding mix. The optimal ratio of the plastic and nonplastic clays is 20–40 and 60–80%, respectively.

The best ratios of the plastic and nonplastic clays, where the molding mixes consisted of low-melting clays from deposits in Lithuania, were determined in [3]. In the present work we investigated the possibility of using a mixture of

low-melting illite-bearing clays to produce frost-resistant brick.

We used a mixture of low-melting clays from the “Ukmyarge” and “Rokai” deposits (80 and 20%, respectively) as well as perlite, whose SiO<sub>2</sub> content was 73%, from the “Fogosh” deposit (TU U 21 V.2.7-526–92), and brick scrap.

The clays used are illite-bearing clays of two types: clay from a Devonian deposit (“Ukmyarge”) and clay from the Quaternary deposit (“Rokai”). These are clays with a narrow roasting interval (30–75°C). The chemical composition of the clays and the perlite are presented in Table 1. The x-ray phase analysis showed that the hydromica, kaolin, quartz, feldspars, dolomite, chlorites, and calcite comprise the crystalline structure of the clays, while amorphous SiO<sub>2</sub>, albite, and quartz comprise the crystalline structure of perlite.

Molding mixes were formulated on the basis of mathematical planning of experiments designed to investigate and optimize the properties of the mixtures (Fig. 1). The minimum and maximum amounts of the raw materials were determined from the published data [4]. After the extreme

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TABLE 1.

Raw material	Content, wt.%							
	SiO <sub>2</sub>	Al <sub>2</sub> O <sub>3</sub>	Fe <sub>2</sub> O <sub>3</sub>	CaO	MgO	Na <sub>2</sub> O	K <sub>2</sub> O	calcination losses
Clay from deposit:								
“Ukmyarge”	66.33	15.80	6.42	1.80	2.72	—	1.63	5.30
“Rokai”	49.81	18.62	7.16	7.35	3.84	—	1.14	12.08
Perlite	72.98	12.92	2.25	0.88	—	2.35	2.82	5.80

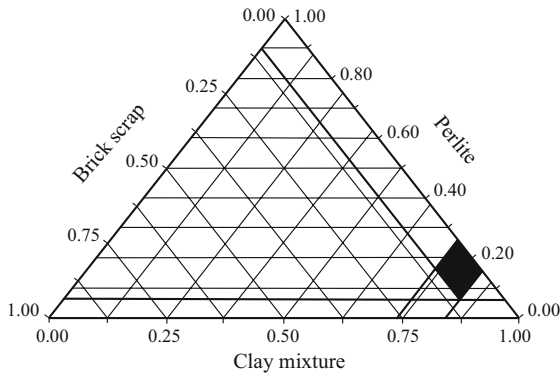


Fig. 1. Triangular diagram for formulating molding mixes.

points were marked in the triangular diagram, a quadrangle was obtained. The vertices and the point of intersection of its diagonals are points indicating the quantitative composition of the molding mixes (Table 2). The clays were dried to constant mass at  $110 \pm 5^\circ\text{C}$ , comminuted in a jaw crusher, and sieved through a No. 1 sieve. The mix prepared for molding was held for 2 h in a moist state.

To determine the main properties,  $70 \times 70 \times 70$ ,  $50 \times 50 \times 50$ , and  $160 \times 30 \times 15$  mm sample were made from the molding mixes by the plastic method. The samples were roasted in a laboratory muffle furnace for 41 h and held for 8 h at  $1070^\circ\text{C}$ . The total shrinkage  $Y_1$  (LST 1272–92), the average density  $Y_2$  (GOST 7025–91), the water absorption  $Y_3$  (GOST 7025–91) with holding for 72 h in water, the bending strength  $Y_4$ , and the compression strength  $Y_5$  (LST 1272–92) as well as the frost resistance  $Y_6$  with omni-directional freezing (GOST 7025–91) were determined for the samples.

Other physical properties of the samples prepared from the molding mixes presented in Fig. 2 are also investigated. The following parameters were determined: water absorption of samples with different holding times in water, the main and developed [3] structural parameters, as well as the frost resistance with uni-directional freezing (GOST 7025–91).

The reliability of the results of parallel tests was checked by means of Student's number with probability 95%. The computed value of Student's number is less than the tabulated value. This shows that the variance of the test results corresponds to the indicated significance. The effect of the quantities of the molding-mix components on the characteristics of the samples can be determined from the values obtained for the coefficients in the regression equations and their absolute values. The least-squares method was used to obtain the regression equations.

Comparing the results obtained for the post-drying and post-roasting shrinkage of the samples obtained using the molding mixes 1–5 (see Table 2) and the mixes prepared only from the experimental clays from the “Ugmyarge” and “Rogoi” deposits in a 4 : 1 ratio, it was established that in the presence of 16–26% addition of perlite and brick scrap the shrinkage of the samples after drying and roasting decreases.

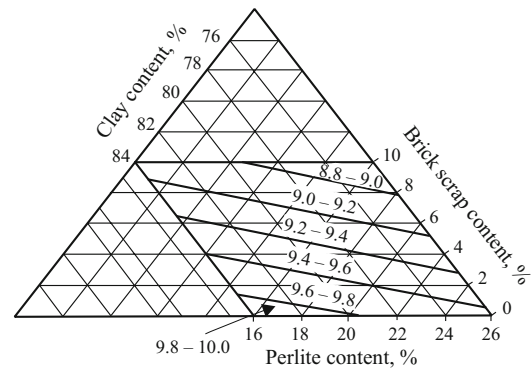


Fig. 2. Distribution of the molding mixes according to the total shrinkage (%). The regions of the molding mixes with total shrinkage from 8.8–9.0 to 9.8–10.0% are shown.

For the samples made only from a mixture of the experimental clays, the shrinkage was 6.18% after drying and 4.6% after roasting. For example, if perlite is introduced in the amount 16% into a molding mix consisting of a mixture of clays (molding mix 4), then the post-drying shrinkage decreases by 1.1% and the total shrinkage decreases by 0.72%.

The following regression equation was obtained to calculate the total shrinkage of the samples:

$$Y_1 = -63.59 + 74.0X_1 + 71.0X_2 + 63.0X_3,$$

where  $X_1$ ,  $X_2$ , and  $X_3$  are the content by weight of the clay mixture, perlite, and brick scrap, respectively.

The multidimensional correlation coefficient of this equation is quite large and equals 0.932; the determination coefficient is 0.868; and, the reproducibility error  $s_e = 0.514\%$ .

The area separated by straight lines according to the total shrinkage of the samples is shown in Fig. 2. Substituting into the equation the component amounts in the molding mix, we obtain the results for total shrinkage of the samples. Evidently, the greater the amount of brick scrap, the lower the total shrinkage of the samples is. The largest total shrinkage of the samples is attained when the molding mix consists of 79.5–84.0% clay mixture, 14.5–20.5% perlite, and 0–1.5% brick scrap.

The density of the samples made from all molding mixes exceeds  $2000 \text{ kg/m}^3$ . The highest density ( $2170 \text{ kg/m}^3$ ) was

TABLE 2.

Molding mix	Content, wt. %			
	clay from “Ugmyarge” deposit	clay from “Rokoi” deposit	perlite	brick scrap
1	59.2	14.8	16	10
2	67.2	16.8	6	10
3	59.2	14.8	26	—
4	67.2	16.8	16	—
5	63.2	15.8	16	5

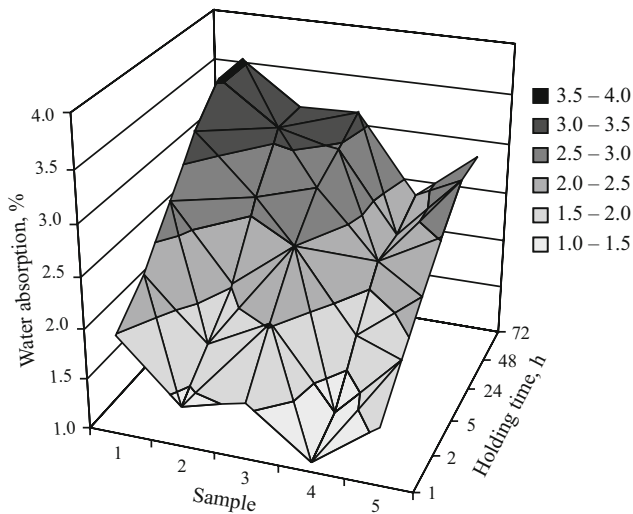


Fig. 3. Water absorption in a sample with different holding times.

obtained for samples made from the molding mix 4. The regression model gives for the average density

$$Y_2 = 6005.14 - 3768.4X_1 - 4171.55X_2 - 4527.0X_3.$$

The multidimensional correlation coefficient of this equation is 0.953, the determination coefficient is 0.909, and  $s_e = 10.49 \text{ kg/m}^3$ .

It is evident from the equation that the amount of perlite has a large effect on the total density. When the amount of perlite is optimal, i.e., 12.5–19.0%, the total density of the samples is 2160–2180  $\text{kg/m}^3$ . The amounts of the other components in the molding mix of the samples are: 81–84% clay mixture and 0–3.5% brick scrap.

The water absorption of the samples after 72 h is less than 5%. The lowest water absorption (2.8%) was observed for samples made from the molding mix 4. The regression model gives for the water absorption after 72 h

$$Y_3 = -201.813 + 204.0X_1 + 207.833X_2 + 216.667X_3.$$

The multidimensional correlation coefficient of this equation is quite large and equals 0.981, the determination coefficient is 0.963, and  $s_e = 0.163\%$ . It is evident from the equation that the amount of perlite has a large effect on the water absorption. The lowest water absorption was recorded for the molding mix consisting of 76.2–84.0% clay mixture, 12.6–23.8% perlite, and 0–3.4% brick scrap.

Water absorption in the samples is not a uniform process. After 1 h the samples absorbed 50% of the total amount of water, and after the first day they absorbed more than 85%. Figure 3 shows how the water absorption in the samples occurred.

In the present work, the structural characteristics of the sample were calculated: the effective porosity according to the water absorption over 72 h  $W_E$ , the total porosity according to the water absorption in vacuum  $W_R$ , the relative available pore space  $R$ , the conditional thickness of the pore and

TABLE 3.

Parameter	Ceramic obtained using the molding mix				
	1	2	3	4	5
$W_E$ , %	7.55	6.65	6.70	5.00	6.07
$W_R$ , %	14.45	14.96	13.71	13.87	13.06
$R$ , %	47.73	55.54	51.10	63.93	53.50
$D$	5.92	5.68	6.30	6.21	6.66
$N$	0.14	0.09	0.17	0.14	0.20
$g_1$ , $\text{g/cm}^2$	0.17	0.09	0.11	0.09	0.13
$g_2$ , $\text{g/cm}^2$	0.15	0.11	0.13	0.07	0.11

capillary walls  $D$ , the degree of structural nonuniformity  $N$ , the mass capillary rate of flow parallel and perpendicular to the direction of freezing  $g_1$  and  $g_2$ , respectively [3]. As expected, the highest relative pore-space availability was obtained for samples made from the molding mix 4 (63.93%). The effective porosity and the mass capillary flow rate  $g_1$  and  $g_2$  were highest for these samples. These samples withstood 125 uni-directional freezing cycles. The average values of the structural parameters are presented in Table 3.

The average bending strength (MPa) was 10.4 for samples made using the first molding mix, 9.0 for the second, 9.3 for the third, 9.3 for the fourth, and 8.7 for the fifth. Comparing the results obtained for the bending strength for the samples made from the molding mixes 1–5 and the mix made using only the experimental clays from the “Ukmyarge” and “Rokai” deposits in the ratio 4 : 1 (bending strength 11.8 MPa), it was determined that when perlite and brick scrap are present in the amounts 16–26%, the bending strength decreases.

The bending strength obtained with the regression model is

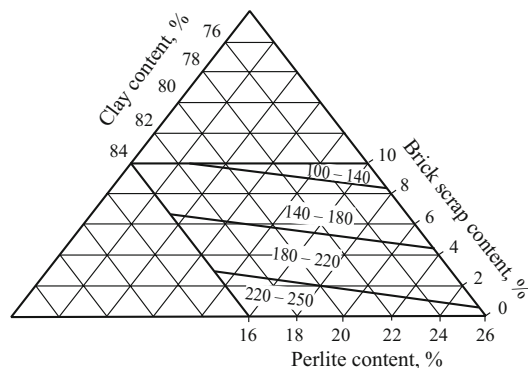
$$Y_4 = 233.588 - 222.6X_1 + 232.9X_2 - 236.0X_3.$$

The multidimensional correlation coefficient of this equation is large and equals 0.993, the determination coefficient is 0.987, and  $s_e = 0.015 \text{ MPa}$ . According to the model the amount of the clay mixture has the greatest effect on the bending strength. According to this equation, the samples made from the molding mix with the following composition have the greatest bending strength (%): 82.7–84.0 clay mixture, 11.5–16.3 perlite, and 0–4.5 brick scrap.

According our regression model the compression strength is

$$Y_5 = 109.861 - 79.4X_1 - 108.467X_2 - 159.333X_3.$$

The multidimensional correlation coefficient of this equation is 0.738, the determination coefficient is 0.545, and  $s_e = 4.452 \text{ MPa}$ . According to the model the amounts of brick scrap and perlite have the greatest effect on the compression strength. The more brick scrap present in the molding mix, the lower the compression strength is. The samples made from the molding mix with the following composition



**Fig. 4.** Distribution of molding mixes over the frost resistance (cycles) for bulk freezing. The regions of molding mixes with frost resistance from 100 – 140 to 220 – 250 cycles are shown.

have the greatest compression strength (%): 81.2 – 84.0 clay mixture, 14.5 – 18.8 perlite, and 0 – 1.5 brick scrap.

The samples were tested for frost resistance with omni-directional freezing. Samples started to break down after 114 cycles for molding mixes 1 and 2 and 193 cycles for mix 3; the samples made from mixes 4 and 5 did not break down after 250 cycles.

The regression model for the frost resistance with omni-directional freezing is

$$Y_6 = -8330.27 + 8620X_1 + 8371.67X_2 + 7333.33X_3.$$

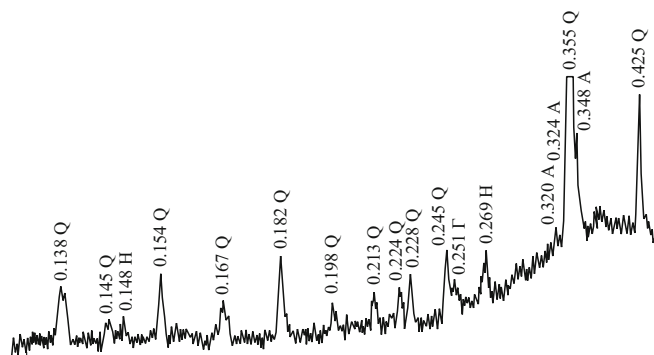
The multidimensional correlation coefficient of this equation is quite large and equals 0.816, the determination coefficient is 0.666, and  $s_e = 7.88$  cycles.

Figure 4 shows the area divided by straight lines with respect to the frost resistance of the samples for omni-directional freezing. The amount of brick scrap has the greatest effect on the frost resistance. If the brick scrap in the molding mix amounts to 3%, then the frost resistance of the samples is 180 – 220 cycles. The samples made from the molding mix with the following composition had the greatest frost resistance (%): 74.0 – 84.0 clay mixture, 13.3 – 26.0 perlite, and 0 – 2.7 brick scrap.

The most widely used method of frost-resistance testing with omni-directional freezing does not completely capture the essence of using ceramic material.

Operational frost resistance characterizes the durability of ceramic materials. According to the existing theoretical ideas, the operational frost resistance can be described as a process of gradual dispersal of a porous body as a result of motion. The destruction is due to the motion of water and plastic ice in the porous material. The structural particularities of the articles determine the resistance to destruction.

At the first stage with uni-directional freezing substantial loads and deformations arise in dangerous, often surface, sections of the sample and spread until the indications of breakdown appear. Surface destruction of the frontal layer of ceramic brick is observed after uni-directional freezing.



**Fig. 5.** X-ray diffraction pattern of the mix 4 roasted at 1070°C (nm): Q) quartz; H) hematite  $\text{Fe}_2\text{O}_3$ ; A) anortite.

The samples made from the molding mixes 1 – 5 were tested under uni-directional freezing. It was determined that the samples made from the mixes 1 and 3 started to break down after 75 cycles, and the samples made from the mixes 2 and 5 started to breakdown after 125 cycles. No breakdown was observed for samples made from the molding mix 4 even after 150 cycles.

In the x-ray diffraction pattern (DRON-2 diffractometer) of the roasted molding mix 4 shown in Fig. 5, the lines attesting to the presence of quartz, hematite  $\text{Fe}_2\text{O}_3$ , and anortite are marked.

It was established on the basis of the experimental data and the calculations performed that frost-resistant ceramic can be obtained from a mixture of low-melting illite-bearing clays without using other, more expensive, high-melting clays.

The optimal composition of the molding mix is as follows (%): 64.96 – 67.20 “Ukmyarge” clay, 16.24 – 16.80 “Rokai” clay, 15 – 19 perlite, 0 – 2 brick scrap. Such mix should be roasted at 1070°C and soaked at constant temperature for 8 h. A sintered ceramic material with the following properties can be obtained under these conditions:

- water absorption with holding time 72 h in water — < 5% (2.5 – 3.5%);
- average density — > 2100 kg/m<sup>3</sup>;
- average compression strength — > 20 MPa; and,
- frost resistance — > 250 cycles for omni-directional freezing and from 75 to 125 cycles for uni-directional freezing.

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